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# [54] SHELL FORMING SYSTEM

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# Related U.S. Application Data

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	1982, abandoned.						

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					5/00

[52] **U.S. Cl.** ...... **264/12**; 65/21.4; 222/1; 222/420; 222/422; 239/8; 239/424; 264/13; 264/574

[56] References Cited

#### U.S. PATENT DOCUMENTS

4,166,084	8/1979	Shea	222/133 X
4,303,431	12/1981	Torobin	264/574 X
4,344,787	8/1982	Beggs et al	65/21.4

#### OTHER PUBLICATIONS

R. J. Calliger et al., "Hollow Drop Production by Injection of Gas Bubbles into a Liquid Jet", *Rev. Sci. Instrum.*, vol. 48, No. 7, 7-1977, pp. 846-851.

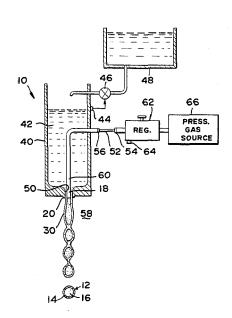
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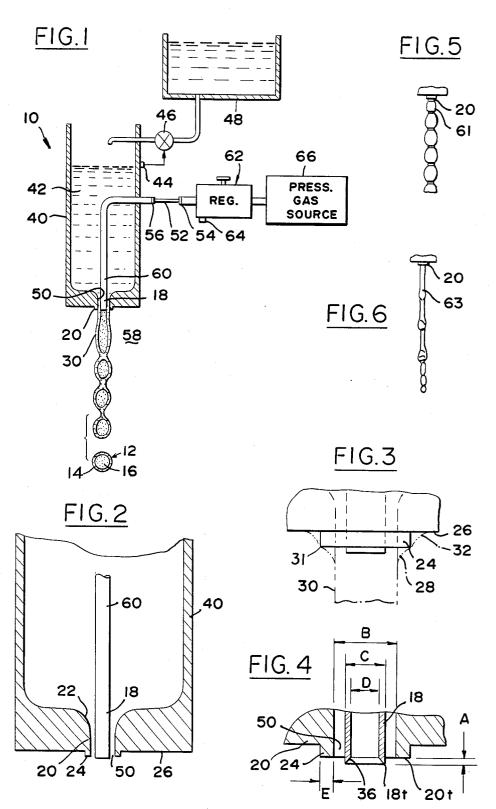
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#### ABSTRACT

An apparatus is provided for forming gas-filled spheres of metal, glass or other material, which produces spheres (12) of uniform size and wall thickness in a relatively simple system. The system includes concentric nozzles, including an inner nozzle (18) through which gas flows and and an outer nozzle (20), which jointly define an annular passageway (50) through which a liquid flows. The flow rates are adjusted so that the gas flows at greater velocity than does the liquid, out of their respective nozzles, e.g. three times as great, in order to produce an extrusion (30) which undergoes axisymmetric oscillations resulting in the pinch off into hollow spheres with very uniform spacing. The system is useful not only where gas-filled spheres are required, but also is useful to accurately control the dispensing of solid, liquid, or gaseous materials.

# 9 Claims, 6 Drawing Figures





#### ORIGIN OF THE INVENTION

SHELL FORMING SYSTEM

The invention described herein was made in the performance of work under a NASA Contract and is subject to the provisions of Section 305 of the National Aeronautics and Space Act of 1958, Public Law 85-568 (72 Stat. 435; 42 USC 2457).

This is a continuation-in-part of application Ser. No. 371,662, filed Apr. 26, 1982, and now abandoned.

#### BACKGROUND OF THE INVENTION

Gas-filled spherical shells can be useful in a variety of 15 applications. For example, very accurately formed shells are required as targets in inertial confinement fusion reactors. A common technique for producing hollow spheres is by the use of concentric nozzles, with gas flowing out of the inner nozzle and molten material 20 flowing through the annular space between the inner and outer nozzles. Various pinch-off mechanisms have been suggested for inducing periodic pinch off of the gas-filled extrusion emerging from the concentric nozzles, as a means for producing hollow spheres of uni- 25 form size. For example, it has been proposed to apply pulses of air transverse to the gas-filled extrusion, or to apply vibrations to the nozzle or to the extrusion emerging from the nozzle to pinch off and break up the extrusion. A simple apparatus and technique which could 30 produce hollow spheres of great uniformity in size and wall thickness, would be of considerable value. If sufficient uniformity could be achieved the apparatus and technique could be useful in precise dispensing of a liquid or gas material.

# SUMMARY OF THE INVENTION

In accordance with one embodiment of the present invention, a relatively simple shell-forming apparatus and method are provided which produce gas-filled shells of high uniformity. The apparatus includes inner and outer nozzles which are concentric, and provides a means for controlling a liquid flow through the annular space between the nozzles, and of a gas through the 45 inner nozzle, in order to produce a hollow pipe-like extrusion emanating from the nozzles. The gas is caused to flow at a higher velocity through the tip of the inner nozzle, than the velocity of liquid at the tip of the outer nozzle, and preferably with a velocity about three times  $_{50}$ as great. It is found that this results in automatic or spontaneous pinch-off of the hollow extrusion into hollow spheres of highly uniform diameter and mass (i.e. average wall thickness), without the need for any additional pinch-off apparatus or technique.

The inner nozzle may extend slightly beyond the outer one, and the outer nozzle may be formed with a protruding lip in order to provide uniform wetting at the tip of the outer nozzle and to enhance uniformity in the breakup of the hollow liquid extrusion.

The novel features of the invention are set forth with particularity in the appended claims. The invention will be best understood from the following description when read in conjunction with the accompanying drawings.

## BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a sectional view of a shell-forming system constructed in accordance with the present invention.

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FIG. 2 is a partial sectional view of the nozzle in the system of FIG. 1.

FIG. 3 is a partial side elevation view of the nozzle of FIG. 2.

FIG. 4 is a partial sectional view of the nozzle of FIG. 3.

FIG. 5 is a side elevation view of an extrusion obtained with the system of FIG. 1 at a low gas-to-liquid velocity ratio.

FIG. 6 is a view similar to that of FIG. 5, but obtained at a high gas-to-liquid ratio.

# DESCRIPTION OF THE PREFERRED EMBODIMENT

FIG. 1 illustrates a system 10 which forms spherical shells 12 that each include a shell wall 14 which is initially liquid and which surrounds a gas core 16. The shells are formed by flowing materials through a nozzle assembly which includes a pair of concentric nozzles 18, 20, with a gas passing out of the inner nozzle 18 and a liquid passing out of the outer nozzle 20. The liquid may be at an elevated temperature such as metal or glass in a molten state, so that the walls of the spherical shells 12 will harden as the shells fall through a tower or other cooling arrangement. A primary goal in the construction and operation of the system is to generate hollow spheres 12 of high uniformity, so that the diameter and average wall thickness (i.e. mass) of the shells are all very close to the same value.

In accordance with the present invention, it is found that shells 12 of uniform size and mass can be formed by controlling the relative velocities of the liquid and gaseous materials at the locations where they issue from their respective nozzles, and in particular by controlling
the velocity of the gaseous material so it is higher than that of the liquid material. The velocity of the gas at the downstream end of its nozzle 18 is preferably about three times that of the liquid at the downstream end of its nozzle 20, that is, the velocity of the gas is preferably
between 1.5 times and 6 times that of the liquid.

Uniformity in shell formation is enhanced by constructing the apparatus to operate in a uniform manner. The inside walls 22(FIG. 2) of the liquid nozzle are gradually curved to direct the liquid out of the nozzle with minimum turbulance. The tip of the outer nozzle may include a lip 24 which projects in front of a surrounding depressed region 26. The lip is used to provide a more uniform meniscus of liquid around the tip of the nozzle to minimize nonuniform effects on the hollow extrusion. As shown in FIG. 3, a meniscus 28 of liquid has been found to frequently form between the lip 24 and the gas-filled liquid extrusion 30 that issues from the nozzle. If a wide flat area surrounded the tip of the nozzle 20, then the size of the meniscus might vary at different locations around the nozzle tip and unsymmetrically affect the extrusion 30. A narrow lip 24 of a wall thickness E (FIG. 4) less than the inside radius of the nozzle 20, but more than one-tenth the radius, avoids this. It may be noted that an additional meniscus 32 may sometimes form between the depressed area 26 and the lip 24, but any irregularity in this auxiliary meniscus 32 has a minimal effect on the extrusion 30, especially since the outer edge 31 of the lip has a sharp corner. The lip is especially useful for liquids that wet the nozzle mate-65 rial.

The uniformity of shell formation is enhanced by avoiding disturbances to the system. In particular, transverse jets of air applied to the extrusion, such as those

used in a bubble blowing method shown in U.S. Pat. No. 4,303,431, are preferably avoided.

The inner nozzle 18 through which gas emerges, is a straight tube of constant diameter. Its tip preferably has an outside diameter at least one-third the inside diame- 5 ter of the outer nozzle. As shown in FIG. 4, the downstream end or tip 18t of the inner nozzle 18 projects a distance A below the tip at 20t of the liquid nozzle. Such slight protrusion of less than one-half the inside radius of the nozzle 20 but more than one-tenth the clearance 10 50 between nozzles, has been found to provide more regular formation of shells. The protrusion A is small, as the tip of the two nozzles must be close to being even with one another. It has also been found that it is preferable to taper the inside of the gas nozzle 18 near its tip, 15 at the location 36, so that the walls of the nozzle 18 are progressively thinner at positions progressively nearer the extreme tip. Such tapering has been found to produce greater uniformity in the shells. It is noted that such tapering has not been used for very small nozzles 20 in applicant's tests, because of the difficulty of fabrica-

The system 10 of FIG. 1 includes a container 40 that holds a quantity of the liquid 42 which is to form the walls of the shells. A predetermined height of the liquid 25 is maintained above the tip of the liquid nozzle 20, in order to maintain a predetermined velocity of liquid out of the nozzle. A sensor 44 senses when the level of the liquid falls belows a predetermined level, to operate a valve 46 that passes additional liquid from a supply 48 30 into the container 40. An alternate scheme has also been used, wherein gas at a predetermined pressure has been maintained against a supply at a low height above the nozzle. In either case, the velocity flow rate can be calibrated by noting the volumetric flow of liquid out of 35 the annular space 50 that lies within the liquid nozzle and outside of the gas nozzle, for a given height of liquid or pressure of gas behind a liquid supply.

The velocity of gas flowing out of the inner nozzle 18 is controlled by passing the gas through a flow-resistant 40 tube 52, which is sometimes referred to as a capillary tube, and by maintaining a predetermined pressure difference between the upstream and downstream ends 54, 56 of the tube. The pressure at the downstream end 56 of the tube is very close to the ambient pressure existing 45 in the area 58 immediately outside the nozzles. This is because the liquid in the hollow stream 30 issuing from the nozzle cannot withstand an appreciable pressure difference between the gas pressure within it and the ambient gas pressure. Also, there is very little pressure 50 drop through the large tube 60 leading from the capillary tube to the nozzle 18. The pressure at the upstream end 54 of the flow-resistant tube 52 is maintained at a predetermined level above the ambient pressure by a pressure regulator 62 which compares the pressure at 55 had outside diameters of about twice the outer nozzle the tube end 54 with the pressure at a regulator inlet 64 that is open to the ambient pressure. The pressured gas is obtained from a high pressure source 66, with the gas passing through the regulator 62 before reaching the flow-resistant tube 52. Where the shells 12 are to con- 60 liquid velocity, and preferably between 1.5 and 6 times tain gas under a high pressure, the area 58 outside the nozzle is enclosed to contain a high pressure, and the regulator inlet 64 is coupled to that environment at 58.

A number of experiments have been conducted to form hollow gas-filled shells in the manner described 65 above. In one experiment, the outer nozzle 20 (FIG. 4) had an inside diameter B of 4.0 mm, and the inner nozzle 18 had an outside diameter of C of 2.5 mm and a con-

near its tip at 36. Thus, the clearance (three-quarter mm) between inner and outer nozzles was about 20% of the diameter of the outer nozzle. A liquid composed of water was flowed out of the annular space at a velocity of 1.09 meters per second, while air was flowed out of the inner nozzle 18 at a velocity 4.2 times as great. The extrusion or flow stream, had the general appearance shown at 30 in FIG. 1, and underwent axisymmetric oscillations that appeared to cause even pinch-off of the extrusion. With the flow rates maintained constant within 0.1%, the frequency of shell formation was con-

stant inside diameter D of about 2 mm which tapered

stant within 0.1% and the weights of the shells were held to approximately the same uniformity. At a higher flow rate of liquid and gas, with the liquid at 4.75 meters per second and gas velocity at 3.0 times as great, substantially the same results occurred, although capillary waves could be observed on the outer surface of the liquid stream.

At ratios of gas-to-liquid flow much higher than 3, the liquid stream did not break into separately pinchedoff bubbles. For example, at a gas-to-liquid velocity ratio of 12.6, a continuous stream was formed with partially formed hollow spheres connected together. Such a stream is shown at 61 in FIG. 5. At a low gas-toliquid flow rate of 1.4, the largely spherical shells were spaced about 5 diameters apart, with a cylindrical stream connecting successive shells, as shown at 63 in FIG. 6. The shells did eventually completely pinch off and break free of the cylinders of liquid flow in front of and behind them, but were not as uniform in size as at a flow ratio of about 3.

In the experiments, it was found that the viscosity of the liquid did not significantly affect the results. In particular, the same effects were achieved for water, as for glycerine which had about 500 times the viscosity of water. The gas composition also did not appear to affect the results, since the gases Freon-12, nitrogen, and helium were also utilized, which span a density range of 30 to 1. Surface tension also did not appear to significantly affect the results, since the results were about the same with distilled water, as with distilled water in which a surficant (Kodak Photo-Flow) was added. The addition of the surficant decreased the frequency of formation by about 1% and increased the diameter of the spheres by about onethird. As a general observation, the addition of surficant improved the apparent quality of the shells, at a station about two meters below the nozzle exit. The 4.0 mm nozzle was operated with gas tubes whose diameters were as small as 1.2 mm. Similar operations as described above were obtained, with the wall thickness of the shells changing according to the clearance between the inner and outer nozzles. In all cases, the shells diameter. Fluid nozzles of various diameters ranging from 0.3 mm to 4.0 mm were used. In all of these trials, good shell formation was obtained for the gas-to-liquid velocities described above (i.e, gas velocity greater than as great).

In the experiments, the velocity of the gas (air) was measured by use of a wet test meter which measures the volumetric flow rate of gas. The pressure of the gas is very close to atmospheric at all locations. It is known that at a small gas flow rate of less than about 10 meters per second, gas is virtually incompressable, where there is no heating of the gas. The velocity of the liquid was 5

measured by measuring the volumetric low rate. The resulting shells has a diameter of about 8 mm.

In one series of tests using the above-described 4.0 mm outside nozzle and 2.5 mm inside nozzle, the liquid velocity was maintained at 1.09 meters per second 5 (m/sec) and the gas was flowed at rates that varied from 1.4 m/sec to 12.6 m/sec (1.3 to 11.6 times liquid velocity), including at a middle velocity of 4.2 m/sec. At a gas velocity of 1.4 m/sec, the stream did break up into shells. It was found that at 1.4 m/sec shells of uniform 10 size were obtained, but at slightly less than 1.4 m/sec velocity the stream did not break up into shells. Shells of uniform size were obtained at greater gas velocities up to about 11 m/sec (10 times liquid velocity). Above a gas velocity of about 11 m/sec the gas-filled stream 15 was formed of a series of connected bubbles; it was found that above an 11 m/sec gas velocity, the bubbles broke before they could separate into individual shells.

Additional tests have been made using nozzle assemblies wherein the inside diameter of the outer nozzle 20 varied between 0.25 mm and 4.0 mm, using a variety of liquids including water, tin, aluminum, gold, lead, and certain plastics. In these tests, the gas velocity was maintained at about three times liquid velocity and very uniform shells were produced. The liquid velocity was a few meters per second.

Applicants did not attempt to use nozzle assemblies larger than 4 mm because they were not able to cool the larger shells so the shells would harden before they touched a solid surface. The smaller shells cool faster so they solidify before they fall onto the bottom of a cooling chamber. In an outer space (zero gravity) environment, larger shells should be readily producable.

Applicant's analysis of the operation of such nozzle assemblies shows that, beside the need to maintain a gas 35 velocity between about 1.3 and 10 times the liquid flow rate, the velocity of liquid emerging from the outer nozzle must exceed a certain rate. The liquid velocity must exceed the velocity of surface tension waves in the liquid material which forms the gasfilled pipe extruded from the nozzle. Otherwise, the first bubble formed immediately outside the nozzle will cling to the nozzle and grow in diameter until it breaks. The surface tension wave velocity  $V_w$  is a function of the thickness D' of the liquid in the gas-filled extrusion (usually the clearance between the inside of the outer nozzle and the outside of the inner nozzle), the density R of the liquid, and the surface tension S of the liquid. They are related by the following equation:

$$V_w = \frac{[2S]^{\frac{1}{2}}}{RD'}$$

where V<sub>w</sub> is approximately the minimum liquid velocity (e.g. cm/sec), S is the surface tension (e.g. dynes/em), R 55 is density (eg gm/cm³), and D' is the clearance between nozzles (e.g. cm). The density R of water is 1 gm/cm³. The surface tension S of pure water is 70 dynes/cm. The nozzle assembly described above had a 4 mm inside diameter for the outer nozzle and a 2.5 mm outside 60 diameter for the inner nozzle, resulting in a clearance of 0.075 cm. For pure water, the minimum liquid speed would be 43 cm/sec. The ratio of surface tension S to density R is about the same for water, tin, aluminum, gold and lead. Thus, for any nozzle having an outer 65 nozzle diameter of 4 mm or less (which is about the greatest diameter shells that can be cooled to a solid before falling onto a support, with presently available

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equipment) and a clearance between inner and outer nozzle of about 20% of the outer nozzle diameter, the liquid velocity must be at least about 43 cm per second. For variable nozzle sizes under these circumstances, the liquid velocity should be at least about  $11/\sqrt{D'}$  cm/sec where D' is the outer nozzle diameter in centimeters.

Although the above-described system was initially developed for applications where gas-filled shells are required, it has great value where the precise dispensing of solid, liquid, or gaseous material is required. The uniformity of droplet mass of about 0.1% or better which has been achieved with a relatively simple system, is orders of magnitude better than has been achieved in simple systems in which solely liquid droplets were formed by the breaking of a solely liquid stream.

Uniform liquid dispersing by a simple system can be useful in a variety of applications, such as in dispensing ink droplets in ink jet printing, dispensing medicine, and dispensing glue. Solid material can be dispensed by using the solidified shells. Gas can be dispensed by allowing liquid shells to break and draining away the liquid.

Thus, the invention provides a system and method for producing gas-filled shells of uniform diameter and wall thickness in a relatively simple manner, and for dispensing material with a high level of uniformity. This is accomplished by passing gas through an inner nozzle and liquid through the annular space between the inner nozzle and an outer nozzle, wherein the velocity of the gas is carefully controlled to be at a level which is greater than that of the liquid but less than ten times as great. The preferred ratio of gas-to-liquid velocity is about 3 times, or in other words, between 1.5 times and 6 times. Shells can be formed over a wider ratio of velocities although at less than about 1.5 times, there is considerable waste of material and the shells are not as uniform. At a ratio of above ten times, the hollow liquid 40 stream does not appear to pinch off into shells. The liquid velocity should be at least about as great as the surface tension wave velocity of the liquid extrusion. The outer nozzle preferably has a lip at its tip, to enhance the uniformity of any meniscus of liquid between the tip and the emerging fluid stream. The inner nozzle is preferably formed with a beveled inner surface at its extreme tip.

Although particular embodiments of the invention have been described and illustrated herein, it is recognized that modifications and variations may readily occur to those skilled in the art and consequently it is intended to cover such modifications and equivalents.

What is claimed is:

1. A method for forming hollow shells, comprising: emitting gas through a nozzle; and

emitting liquid through an annular area surrounding said nozzle to form a hollow extrusion;

said steps of emitting including moving said gas out of said nozzle at a velocity which is between about 1.3 and 10 times greater than the velocity of emission of said liquid through said annular area, whereby to enable spontaneous pinch-off of said hollow extrusion into said hollow shells.

2. The method described in claim 1 wherein:

the velocity V in centimeters of the liquid through said annular area is at least equal to  $11/\sqrt{D}$  where D is the thickness of said annular area in centimeters.

3. A method for dispensing droplets of initially liquid material, of uniform mass, comprising:

emitting a gas through an inner nozzle; and emitting said liquid material through an annular space that lies between said inner nozzle and an outer nozzle that surrounds said inner nozzle to form a hollow extrusion;

said steps of emitting including moving said gas through the downstream end of said inner nozzle at a velocity which is between about 1.3 and 10 times greater than the velocity of emission of said liquid through the downstream end of said annular space, and controlling the environment in front of said nozzles so it is substantially free of winds that 15 would deflect the emitted gas and liquid sidewardly, whereby to enable spontaneous pinch-off of said hollow extrusion into hollow spheres.

4. The method described in claim 3 wherein: said step of emitting said gas includes emitting said gas from the tip of the inner nozzle into the surrounding environment at a location forward of the location where said liquid material is emitted.

5. A method for dispensing droplets of initially liquid 25 material, of uniform mass, comprising:

emitting a gas through an inner nozzle; and emitting said liquid material through an annular space that lies between said inner nozzle and an outer nozzle that surrounds said inner nozzle;

said steps of emitting including moving said gas through the downstream end of said inner nozzle at a velocity which is between about 1.3 and 10 times greater than the velocity of emission of said liquid through the downstream end of said annular space, and controlling the environment in front of said nozzles so it is substantially free of winds that would deflect that emitted gas and liquid sidewardly;

the velocity of said liquid is at least about as great as the velocity V given by the following equation:

 $V = [2S/RD]_{2}^{1}$ 

where V is the velocity in cm/sec, S is the surface tension of the liquid in dynes/cm, R is the density

of the liquid in gm/cm<sup>3</sup>, and D is the clearance between said inner and outer nozzles in cm.

6. A method for forming hollow shells, comprising: emitting gas through an inner nozzle; and

emitting liquid through an annular area between an outer nozzle that surrounds said inner nozzle and said inner nozzle to form a hollow extrusion;

said steps of emitting including moving said gas out of said inner nozzle at a velocity which is between about 1.3 and 10 times greater than the velocity of emission of said liquid through said annular area;

said outer nozzle has an inside diameter on the order of 4 millimeters, the clearance between said nozzles is on the order of three-quarters millimeter, and the velocity of the liquid is on the order of one meter per second, whereby to enable spontaneous pinch-off of said hollow extrusion into said hollow shells.

7. A method for forming hollow shells, comprising: emitting gas through a nozzle; and

emitting liquid through an annular area surrounding said nozzle to form a hollow extrusion;

said steps of emitting including moving said gas out of said nozzle at a velocity which is between 1.3 and 10 times greater than the velocity of emission of said liquid through said annular area;

the velocity of said liquid which is emitted from said annular area being greater than the velocity of surface tension waves in the liquid material which surrounds the gas in the gas-filled liquid extrusion, whereby to enable spontaneous pinch-off of said hollow extrusion into said hollow shells.

8. The method described in claim 7 wherein:

the velocity V of said liquid which is emitted from said annular area is at least about as great as that given by the following equation:

 $V=[2S/RD]\frac{1}{2}$ 

where V is the liquid velocity in cm/sec, S is the surface tension of the liquid in dynes/cm, R is the density of the liquid in gm/cm<sup>3</sup>, and D is the thickness of said annular area in cm.

9. The method described in claim 7 wherein:

said steps of emitting include moving said gas out of said nozzle at a velocity about three times the velocity of the liquid through the annular space.

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